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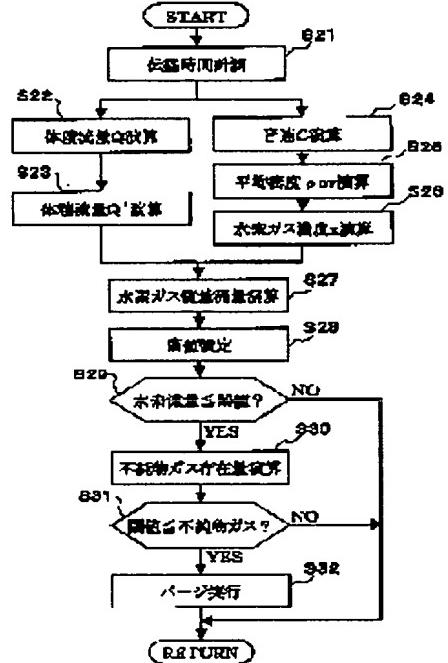
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## (54) FUEL CELL SYSTEM AND CONTROL METHOD

### (57)Abstract:

**PROBLEM TO BE SOLVED:** To improve energy efficiency of a fuel cell system by reducing the undesired loss of fuel gas by purging in the fuel cell system.

**SOLUTION:** Volume flow rate is calculated based on a propagating time of ultrasonic wave in mixed gas measured by an ultrasonic flowmeter, and an average density of the mixed gas is determined. Hydrogen gas concentration and impurity gas concentration are calculated based on the average density, the mass flow rate of hydrogen gas is calculated based on the hydrogen gas concentration and the volume flow rate, and an existing amount of the impurity gas is calculated based on the impurity gas concentration, pressure, and temperature. Here, when the mass flow rate of hydrogen is at a threshold or lower and the existing amount of the impurity gas is at a threshold or higher, the purge is performed.



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**CLAIMS**

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**[Claim(s)]**

[Claim 1] The gas purge valve which is the fuel cell system equipped with the hydrogen reflux path of making the hydrogen which was not used for the generation of electrical energy in the fuel cell flowing back to the path which supplies hydrogen to said fuel cell, and is prepared in said hydrogen reflux path, The fuel cell system characterized by being constituted including a hydrogen mass flow rate presumption means to presume the hydrogen mass flow rate within the hydrogen circulatory system including said hydrogen reflux path, and the purge control means which opens said gas purge valve when said hydrogen mass flow rate becomes below a threshold.

[Claim 2] The fuel cell system according to claim 1 which it has an impurity gas abundance presumption means to presume the impurity gas abundance in the mixed gas within said hydrogen circulatory system, and said purge control means is [ said hydrogen mass flow rate ] below a threshold, and is characterized by opening said gas purge valve when said impurity gas abundance becomes beyond a threshold.

[Claim 3] The fuel cell system according to claim 1 or 2 characterized by establishing a threshold setting means to set up the threshold of said hydrogen mass flow rate according to the service condition of said fuel cell.

[Claim 4] The fuel cell system according to claim 3 by which said threshold setting means is characterized by setting up the threshold of said hydrogen mass flow rate according to the generation-of-electrical-energy current as said service condition.

[Claim 5] While searching for the acoustic velocity in the mixed gas within said hydrogen circulatory system, said hydrogen mass flow rate presumption means and/or an impurity gas abundance presumption means Ask for the mean density of said mixed gas based on this acoustic velocity, and it is constituted including a concentration presumption means to presume the hydrogen concentration and/or impurity gas concentration in said mixed gas based on this mean density. The fuel cell system of any one publication of claim 1-4 characterized by presuming a hydrogen mass flow rate and/or impurity gas abundance based on the concentration called for by this concentration presumption means.

[Claim 6] The fuel cell system according to claim 5 characterized by constituting said hydrogen mass flow rate presumption means including a volumetric flow rate detection means to detect the volumetric flow rate of said mixed gas based on the propagation time of the supersonic wave in the mixed gas within said hydrogen circulatory system , and for said concentration presumption means searching for acoustic velocity based on the measurement result of the propagation time by said volumetric flow rate detection means , and presuming said hydrogen concentration and/or impurity gas concentration .

[Claim 7] A volumetric flow rate detection means to detect the volumetric flow rate of said mixed gas based on the propagation time of the supersonic wave in the mixed gas within said hydrogen circulatory system, While searching for the acoustic velocity in said mixed gas based on a pressure detection means to detect the pressure of said mixed gas, a temperature detection means to detect the temperature of said mixed gas, and the propagation time of said supersonic wave A concentration presumption means to ask for the mean density of said mixed gas based on this acoustic velocity, and to presume the hydrogen gas concentration and impurity gas concentration in said mixed gas based on this mean density, A preparation and said hydrogen mass flow rate presumption means presume a hydrogen mass flow rate based on said volumetric flow rate, a pressure, temperature, and hydrogen gas concentration. The fuel cell system according to claim 2 by which said impurity gas abundance presumption means is characterized by presuming said impurity gas abundance based on said pressure, temperature, and impurity gas concentration.

[Claim 8] The fuel cell system of any one publication of claim 5-7 by which a dehumidification means is infix in said hydrogen reflux path, and said concentration presumption means is characterized by

presuming hydrogen concentration and/or impurity gas concentration based on the propagation time of the supersonic wave in the downstream of said dehumidification means.

[Claim 9] The fuel cell system of any one publication of claim 5-7 by which a humidity detection means to detect the humidity of said mixed gas is established near the measurement part of the propagation time of said supersonic wave, and said concentration presumption means is characterized by presuming said hydrogen gas concentration and/or impurity gas concentration based on the steam concentration called for based on said humidity, and said mean density.

[Claim 10] The fuel cell system of any one publication of claim 5-7 characterized by to establish a temperature detection means detect the temperature of a pressure detection means to detect the pressure of said mixed gas , and mixed gas , near the measurement part of the propagation time of said supersonic wave , and for said concentration presumption means to ask for the steam concentration in a saturated steam condition from said pressure and temperature , and to presume said hydrogen gas concentration and/or impurity gas concentration based on this steam concentration and said mean density .

[Claim 11] The fuel cell system of any one publication of claim 5-7 which said concentration presumption means considers as the constant value which was able to define the steam concentration of said mixed gas beforehand, and is characterized by presuming said hydrogen gas concentration and/or impurity gas concentration based on this steam concentration and said mean density.

[Claim 12] The control approach of the fuel cell system characterized by to make mixed gas purge from said hydrogen reflux path when the hydrogen mass flow rate within the hydrogen circulatory system including said hydrogen reflux path presumes in the fuel cell system equipped with the hydrogen reflux path made to flow back to the path which supplies hydrogen for the hydrogen which was not used for the generation of electrical energy in the fuel cell to said fuel cell and said hydrogen mass flow rate becomes below a threshold.

[Claim 13] In the fuel cell system equipped with the hydrogen reflux path of making the hydrogen which was not used for the generation of electrical energy in the fuel cell flowing back to the path which supplies hydrogen to said fuel cell The hydrogen mass flow rate within the hydrogen circulatory system including said hydrogen reflux path is presumed, and the impurity gas abundance in the mixed gas within said hydrogen circulatory system is presumed. Said hydrogen mass flow rate below with a threshold And the control approach of the fuel cell system characterized by making mixed gas purge from said hydrogen reflux path when said impurity gas abundance becomes beyond a threshold.

[Claim 14] The control approach of the fuel cell system according to claim 12 or 13 characterized by setting up the threshold of said hydrogen mass flow rate according to the generation-of-electrical-energy current of said fuel cell.

[Claim 15] The control approach of the fuel cell system any one publication of claim 12-14 characterize by search for the acoustic velocity in said mixed gas , ask for the mean density of said mixed gas based on this acoustic velocity based on the propagation time of the supersonic wave in the mixed gas within said hydrogen circulatory system , presume the hydrogen concentration and/or impurity gas concentration in said mixed gas based on this mean density , and presume a hydrogen mass flow rate and/or impurity gas abundance based on said concentration .

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**DETAILED DESCRIPTION**

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**[Detailed Description of the Invention]**

[0001]

[Field of the Invention] This invention relates to the technique which controls the purge from a reflux system in the fuel cell system of the type which circulate through fuel gas, and a fuel cell is made to flow back in detail about a fuel cell system and the control approach, and is reused.

[0002]

[Description of the Prior Art] Flowing back the hydrogen which was not consumed with a fuel cell to a fuel cell in a fuel cell system, and aiming at reuse from the former, is performed (refer to JP,10-055814,A and JP,2000-58092,A). Moreover, in the case of the system which fuel gas is made to flow back as mentioned above, and is reused, it is known that impurity gas (for example, nitrogen gas unnecessary for a generation-of-electrical-energy operation of a fuel cell) will accumulate and increase gradually into a circulation line as it continues circulating through fuel gas.

[0003] Continuing circulating such unnecessary gas will cause the loss of power of a fuel cell because the rate of impurity gas of it not only using circulation power vainly, but occupying it in the fuel supplied to a fuel cell increases. Then, it has the purge valve which makes the fuel gas which contains impurity gas from the circulation line of fuel gas purge, and when the impurity gas concentration in a circulation line exceeds constant value, said purge valve is made to open wide with the configuration indicated by said JP,2000-058092,A.

[0004]

[Problem(s) to be Solved by the Invention] By the way, in the fuel cell system which fuel gas is made to flow back and is reused, if the hydrogen supply flow rate from a fuel gas source of supply is set to  $Q$  (L/min), to this  $Q$  (L/min), flow rate  $\alpha Q$  (L/min) of a certain ratio  $\alpha$  will be absorbed, and will flow back with an ejector, and  $Q(L/min)$  which is a sum total flow rate ( $1+\alpha$ ) will flow into a fuel cell.

[0005] And it has a relation that  $\alpha Q$  (L/min) which remained without consuming and consuming the hydrogen of  $Q$  (L/min) with a fuel cell is discharged from a fuel cell as unused gas, and circulates. That is, the hydrogen flow  $Q$  (L/min) which flows from a fuel gas source of supply is consumed with a fuel cell, and amount  $\alpha Q$  (L/min) depending on this flow  $Q$  (L/min) circulates.

[0006] It is required from the amount of [ for the hydrogen mass flow rate which flows into a fuel cell so that it may be indicated by JP,2000-012059,A being related to the generating efficiency of a fuel cell, and making the amount more than the hydrogen mass flow rate consumed with a fuel cell flow into a fuel cell being searched for, and on the other hand, corresponding to rapid load increase of a fuel cell ] allowances being need that the hydrogen mass flow rate through which it circulates should be more than predetermined.

[0007] Since there is no effect in a generation of electrical energy of a fuel cell, with [ the hydrogen mass flow rate (a part for allowances) through which it will circulate if it puts in another way ] predetermined [ more than ], the purge from a circulation line, but With the configuration which controls a purge based on impurity gas concentration, like before Since it was not able to respond to the hydrogen mass flow rate in a circulation line changing with the consumption in a fuel cell with the same impurity gas concentration but, there was a problem of the ability not to make it purge to suitable timing.

[0008] Moreover, with the configuration which controls a purge based on impurity gas concentration like before, it is influenced by change of transitional impurity gas concentration, a purge is performed, and loss of unnecessary fuel gas may have been increased. For example, if there is increase change of the load of a fuel cell, since the hydrogen mass of a circulation line will fall temporarily and impurity gas concentration will increase relatively by it in the condition with the impurity gas of a circulation line that amount are recording is carried out, in the conventional control, a purge may be performed based on increase of applied

temporary impurity gas concentration.

[0009] However, since to refrain from a purge was desired when the are recording abundance of impurity gas is below the specified quantity, considering the viewpoint of performing an effective purge, in the conventional control, loss of unnecessary fuel gas may have been increased by frequent purge control. This invention is made in view of the above-mentioned trouble, and it aims at offering the fuel cell system which unnecessary loss of fuel gas is reduced and can raise the energy efficiency of a fuel cell system, and the control approach.

[0010]

[Means for Solving the Problem] In order to attain the above-mentioned purpose, when the hydrogen mass flow rate within the hydrogen circulatory system including a hydrogen reflux path was presumed and said hydrogen mass flow rate became below a threshold, by invention of claim 1 and 12 publications, it considered as the configuration which makes mixed gas purge from a hydrogen reflux path. When the hydrogen mass flow rate which changes according to flow rate change of the mixed gas within the hydrogen circulatory system (hydrogen gas, impurity gas, steam) according to the above-mentioned configuration becomes below a threshold and it becomes impossible to maintain generating efficiency, it purges and impurity gas is made to purge with fuel gas.

[0011] In invention of claim 2 and 13 publications, the hydrogen mass flow rate and impurity gas abundance within the hydrogen circulatory system were presumed, respectively, said hydrogen mass flow rate was below a threshold, and when said impurity gas abundance became beyond a threshold, it considered as the configuration which makes mixed gas purge from a hydrogen reflux path. According to the above-mentioned configuration, even if a hydrogen mass flow rate becomes below a threshold by the change of load of a fuel cell etc., a purge will not be performed when impurity gas abundance is less than a threshold.

[0012] In invention according to claim 3, it considered as the configuration which sets up the threshold of said hydrogen mass flow rate according to the service condition of said fuel cell. According to the above-mentioned configuration, the threshold of the hydrogen mass flow rate which makes it purge so that the hydrogen mass flow rate needed may make it respond to changing according to the service condition of a fuel cell is changed.

[0013] In invention of claim 4 and 14 publications, it considered as the configuration which sets up the threshold of said hydrogen mass flow rate according to the generation-of-electrical-energy current of a fuel cell. According to the above-mentioned configuration, the threshold of a hydrogen mass flow rate is changed corresponding to the hydrogen mass flow rate needed according to a generation-of-electrical-energy current for maintenance of generating efficiency changing. In invention of claim 5 and 15 publications, it considered as the configuration which makes the hydrogen concentration and/or impurity gas concentration which are used for presumption of a hydrogen mass flow rate and/or impurity gas abundance presume based on the mean density of mixed gas asked based on the acoustic velocity in the mixed gas within the hydrogen circulatory system.

[0014] When according to the above-mentioned configuration the acoustic velocity in mixed gas shows the mean density of mixed gas, for example, mixed gas consists of hydrogen and impurity gas, the sum total of hydrogen concentration and impurity gas concentration is 1, and since the result of having added the multiplication value of the concentration of each gas and a consistency serves as mean density, it is possible to ask for hydrogen concentration and impurity gas concentration based on these formulas.

[0015] In invention according to claim 6, it considered as the configuration using a means to detect a volumetric flow rate to presumption of a hydrogen mass flow rate based on the propagation time of the supersonic wave in the mixed gas within the hydrogen circulatory system, and considered as the configuration using the measurement result of the propagation time for the detection of said volumetric flow rate to concentration presumption. According to the above-mentioned configuration, based on the propagation time measured for detection of a volumetric flow rate, presumption of hydrogen concentration and/or impurity gas concentration is performed to coincidence.

[0016] A means to detect a volumetric flow rate in invention according to claim 7 based on the propagation time of a supersonic wave, A means to detect the pressure of mixed gas, and a means to detect the temperature of mixed gas, Based on the mean density which searches for the acoustic velocity in mixed gas based on a preparation and said propagation time, and is called for based on this acoustic velocity, hydrogen gas concentration and impurity gas concentration are presumed. While presuming the hydrogen mass flow rate based on said volumetric flow rate, a pressure, temperature, and hydrogen gas concentration, it considered as the configuration which presumes impurity gas abundance based on said pressure, temperature, and impurity gas concentration.

[0017] According to the above-mentioned configuration, concentration presumption is performed with detection of a volumetric flow rate, and a hydrogen mass flow rate and impurity gas abundance are presumed corresponding to change of a pressure and temperature. In invention according to claim 8, it considered as the configuration which makes hydrogen concentration and/or impurity gas concentration presume based on the propagation time in the downstream of a dehumidification means. According to the above-mentioned configuration, when the mixed gas of the hydrogen circulatory system consists of hydrogen, impurity gas, and a steam, by being dehumidified by the upstream of the measurement part of the propagation time, mixed gas will consist of two components of hydrogen and impurity gas, and an average with the consistency of hydrogen and the consistency of impurity gas will be called for from the propagation time. Therefore, an unknown serves as only hydrogen concentration and impurity gas concentration, and hydrogen concentration and impurity gas concentration will be called for from the equation of concentration, and the equation of a consistency.

[0018] In invention according to claim 9, humidity was detected near the measurement part of the propagation time, and it considered as the configuration which asks from the steam concentration and the propagation time which are found based on this humidity, and presumes hydrogen gas concentration and/or impurity gas concentration based on \*\*\*\* mean density. According to the above-mentioned configuration, even if mean density is the configuration called for as mean density of hydrogen, impurity gas, and a steam, hydrogen concentration and impurity gas concentration will be called for by making concentration of a steam into known.

[0019] In invention according to claim 10, near the measurement part of the propagation time, a pressure and temperature were detected, and it asked for the steam concentration in a saturated steam condition from said pressure and temperature, and considered as the configuration which presumes hydrogen gas concentration and/or impurity gas concentration based on this steam concentration and the mean density called for from the propagation time. According to the above-mentioned configuration, if the inside of the hydrogen circulatory system is in an abbreviation saturated steam condition, steam concentration will be called for from a pressure and temperature, and hydrogen concentration and impurity gas concentration will be called for by making concentration of a steam into known.

[0020] In invention according to claim 11, it considered as the constant value which was able to define the steam concentration of mixed gas beforehand, and considered as the configuration which presumes hydrogen gas concentration and/or impurity gas concentration based on this steam concentration and the mean density called for from the propagation time. According to the above-mentioned configuration, hydrogen concentration and impurity gas concentration are called for by assuming the concentration of a steam to be constant value.

[0021]

[Effect of the Invention] a purge is performed in the condition that the hydrogen mass flow rate which can maintain generating efficiency is secured since a purge is controlled based on the hydrogen mass flow rate in the hydrogen circulatory system according to invention of claim 1 and 12 publications -- avoidable -- with -- \*\*\* -- it is effective in the ability to reduce loss of the hydrogen gas by unnecessary purge.

[0022] Since according to invention of claim 2 and 13 publications a purge is not performed when there is little impurity gas abundance even if a hydrogen mass flow rate falls When a hydrogen mass flow rate falls transitionally, can avoid that a purge will be performed, although there is little impurity gas abundance which should be purged, and only an effective purge is made to perform, and it is effective in the ability to reduce loss of the hydrogen gas by much more unnecessary purge.

[0023] Since it is made to purge based on whether it has a hydrogen mass flow rate which can maintain generating efficiency and can secure a part only for the allowances which can respond to a load effect according to invention of claims 3 and 4 and 14 publications, there is effectiveness of the ability to make it purge to the always optimal timing according to change of the service condition (load) of a fuel cell. Since hydrogen concentration and/or impurity gas concentration are presumed from the acoustic velocity in mixed gas according to invention of claim 5 and 15 publications, it is effective in that concentration information required for presumption of a hydrogen mass flow rate and impurity gas abundance may be based on acoustic velocity.

[0024] According to invention according to claim 6, concentration information can be acquired to coincidence with detection of the volumetric flow rate for presuming a hydrogen mass flow rate, and there is effectiveness of the ability to make a hydrogen mass flow rate and impurity gas abundance presume with a simple configuration. According to invention according to claim 7, concentration information can be acquired to coincidence with detection of the volumetric flow rate for presuming a hydrogen mass flow rate,

and it is effective in the ability to presume a hydrogen mass flow rate and impurity gas abundance with high precision corresponding to change of the pressure within the hydrogen circulatory system, and temperature. [0025] It is effective in the ability to presume hydrogen concentration and impurity gas concentration, without performing density measurement of steam gas, since the propagation time in the mixed gas after dehumidification is made to measure in the configuration which makes hydrogen concentration and/or impurity gas concentration presume based on the propagation time of the supersonic wave in mixed gas according to invention according to claim 8. According to invention according to claim 9, there is effectiveness of the ability to make hydrogen concentration and impurity gas concentration presume from the propagation time of the supersonic wave in the mixed gas containing a steam by asking for steam concentration from the detection result of humidity.

[0026] According to invention according to claim 10, when the inside of the hydrogen circulatory system is in a saturated steam condition, it asks for steam concentration simple from a pressure and temperature, and there is effectiveness of the ability to make hydrogen concentration and impurity gas concentration presume from the propagation time of the supersonic wave in the mixed gas containing a steam. When the humidity within the hydrogen circulatory system, a pressure, and temperature are under the condition it can be considered that is abbreviation regularity according to invention according to claim 11, there is effectiveness of the ability to make hydrogen concentration and impurity gas concentration presume simple from the propagation time of the supersonic wave in the mixed gas containing a steam.

[0027]

[Embodiment of the Invention] The gestalt of operation of this invention is explained based on drawing below. Drawing 1 is drawing showing the fuel cell system in an operation gestalt. In this drawing 1, the hydrogen gas from the source 1 of hydrogen gas supply is introduced into the fuel electrode (illustration abbreviation) of the fuel cell stack 20 through the supply pressure regulation bulb 2, hydrogen gas supply Rhine 3, an ejector 4, and hydrogen gas supply Rhine 5, and the fuel gas which was not consumed by the fuel cell stack 20 flows back to hydrogen gas supply Rhine 5 with said ejector 4 through the hydrogen gas reflux ways 7, 9, and 10 (hydrogen reflux path).

[0028] The hydrogen which was not used for the generation of electrical energy in the fuel cell stack 20 by the above-mentioned configuration flows back to a fuel cell 20 again, and the circulatory system of hydrogen is constituted by the hydrogen gas reflux ways 7, 9, and 10 and hydrogen gas supply Rhine 5. The purge valve 11 is infixed in discharge Rhine 12 by which branching installation is carried out from said hydrogen gas reflux way 7.

[0029] The dehumidifier 8 (dehumidification means), the thermometer 37 (temperature detection means), the flowmeter 30 (volumetric flow rate detection means), and the pressure gage 36 (pressure detection means) are infixed in the hydrogen gas reflux way 9 of the downstream sequentially from the upstream rather than the tee of said discharge Rhine 12. Moreover, the humidifier 6 is infixed in said hydrogen gas supply Rhine 5. The signal data-processing section 40 controls disconnection of said purge valve 11 based on data processing which inputs and mentions the detecting signal from said thermometer 37, a flowmeter 30, and a manometer 36 later.

[0030] Drawing 2 shows the detail of said flowmeter 30. Said flowmeter 30 is a supersonic type flow meter in this operation gestalt, and is equipped with the ultrasonic reception-and-transmission machines 31 and 32. The acoustic wave aslant emitted towards the downstream from the ultrasonic reception-and-transmission machine 31 of the upstream Reflect in the ultrasonic reflective section 33 and it is received by the ultrasonic reception-and-transmission machine 32 of the downstream. The propagation time  $t_1$  of the supersonic wave which the acoustic wave aslant emitted towards the upstream from the ultrasonic reception-and-transmission machine 32 of the downstream reflects in the ultrasonic reflective section 33, is received by the ultrasonic reception-and-transmission machine 31 of the upstream, and goes to the downstream, The propagation time  $t_2$  of the supersonic wave which goes to the upstream is measured.

[0031] Here, when mean velocity in L and the hydrogen gas reflux way 9 is set [ the include angle of the axis of the hydrogen gas reflux way 9, and a supersonic wave / the acoustic velocity of theta and gas ] to V for the die length of the propagation path 34 of C and a supersonic wave, said propagation times  $t_1$  and  $t_2$  (sec) are  $t_1=L/(C+V\cos\theta)$ . .... (1)

$$t_2=L/(C-V\cos\theta) \quad \dots \quad (2)$$

If it comes out, and it is expressed and mean velocity V (m/s) and acoustic velocity C (m/s) are searched for from the two above-mentioned formulas  $V=L/(2\cos\theta) * (1/t_1-1/t_2)$  .... (3)

$$C=L/2 * (1/t_1+1/t_2) \quad \dots \quad (4)$$

It becomes.

[0032] Therefore, the average volumetric flow rate Q in the hydrogen gas reflux way 9 (m<sup>3</sup>/s) is  $Q=V \cdot S \cdot K$ , when the duct cross section is set to S and it sets a flow rate correction factor to K. .... (5)  
 Although it comes out and asks, since said average volumetric flow rate Q is a volumetric flow rate in reference temperature and a reference pressure condition, it computes Q' (normal cube meter / second) by performing amendment according to the pressure p and temperature T at that time.

[0033]

$$Q'(Nm^3/s) = Qx(p/p_0) \times (T_0/T) \dots (6)$$

In addition, by the upper formula, p<sub>0</sub> shows reference pressure and T<sub>0</sub> shows reference temperature. When it has composition which controls a purge by this operation gestalt based on the hydrogen mass flow rates dmH<sub>2</sub>/dt, and hydrogen gas concentration is set to x and hydrogen density is set to rho x, the hydrogen mass flow rates dmH<sub>2</sub>/dt (kg/s) are dmH<sub>2</sub>/dt=x-rho x-Q'. .... (7)

It is computed by carrying out (hydrogen mass flow rate presumption means).

[0034] Here, although hydrogen, a steam, and the mixed gas that consists of impurity gas (nitrogen) flow to the circulatory system, since said ultrasonic-type flowmeter 30 is arranged in the downstream of a dehumidifier 8, said acoustic velocity C turns into the acoustic velocity C in the mixed gas which consists of two components of the hydrogen and impurity gas (nitrogen) except a steam. Moreover, when the component of the mixed gas which searched for acoustic velocity C is known, acoustic velocity C is expressed with the function of the mixing ratio (concentration) of each component (refer to JP,2000-304732,A), and becomes correlation as indicated to be mean density rho<sub>av</sub> and acoustic velocity C of the starting mixed gas to drawing 3 in the case of two components of hydrogen and impurity gas (nitrogen).

[0035] Therefore, if the acoustic velocity C in mixed gas is known, mean density rho<sub>av</sub> of mixed gas can be found according to drawing 3, and it can ask for the concentration of each component from mean density rho<sub>av</sub>. That is, when the consistency of x and hydrogen gas is set to rho x for the concentration of hydrogen gas and the consistency of y and impurity gas is set to rho y for the concentration of impurity gas, since it consists of two components of hydrogen gas and impurity gas, mixed gas is x+y=1. .... (8)

$$X-rho x+y-rho y=rho av \dots (9)$$

$$Since come out and it is, it is x=(rho y-rho av)/(rho y-rho x). \dots (10)$$

$$y=(rho av-rho x)/(rho y-rho x) \dots (11)$$

Mean density rho<sub>av</sub> to consistency rho[ of the consistency rho x of a next door and hydrogen gas and impurity gas ] y, and the hydrogen gas concentration x and impurity gas concentration y calculated from acoustic velocity C are computable (concentration presumption means).

[0036] In addition, consistency rho[ of the consistency rho x of the hydrogen gas used when asking for the concentration x of the above-mentioned hydrogen gas and impurity gas concentration y, and impurity gas ] y is set up from the pressure and temperature at that time. carrying out the multiplication of said volumetric flow rate Q' to the applied concentration x and y, if the hydrogen gas concentration x and impurity gas concentration y are called for as mentioned above -- hydrogen -- atmosphere -- product flow rate Qx' and an impurity -- atmosphere -- product flow rate Qy' is called for and a hydrogen mass flow rate and an impurity gas mass flow rate are calculated by carrying out the multiplication of the gas consistency rho x and the rho y to this volumetric flow rate Qx' and Qy'.

[0037] Since the hydrogen mass flow rate supplied to a fuel cell affects the generating efficiency of a fuel cell and the amount of [ for making the amount more than the hydrogen mass flow rate consumed with a fuel cell flow into a fuel cell being searched for, and corresponding to rapid load increase of a fuel cell ] allowances are also need, it is required that the amount of [ which is added to a part for the hydrogen mass flow rate in a circulation line, i.e., consumption, ] allowances should be more than predetermined.

[0038] On the other hand, since the flow rate of a certain ratio flows back automatically with an ejector 4, when a reflux quantity of gas flow has few hydrogen mass flow rates in reflux gas than the amount corresponding to the above-mentioned demand, required hydrogen capacity will run short and unnecessary gas constituents (impurity gas) will be conversely filled with a fuel cell. So, in said signal data-processing section 40, if the hydrogen mass flow rate and threshold which were computed as mentioned above are compared and a hydrogen mass flow rate becomes below a threshold, the impurity gas which opened the purge valve 11 and collected in the circulatory system will be purged until it will be in the condition that the required amount of hydrogen is securable with a fuel cell (for example, fixed time amount) (purge control means).

[0039] Here, when it is dependent on the operational status of a fuel cell and considers that a fuel cell is a converter, as for the hydrogen gas mass flow rate required for a fuel cell, it is rational to set it as the amount corresponding to the generation-of-electrical-energy current value which is the output, and it is good to

change said threshold according to a generation-of-electrical-energy current (threshold setting means). If said hydrogen gas concentration x and volumetric flow rate Q' are made into a variable as shown in drawing 4 If the case where the condition of hydrogen mass flow rate regularity is expressed with a hyperbola group, and it operates steadily with the hydrogen mass flow rate a corresponding to a certain generation-of-electrical-energy current value is assumed, as shown in drawing 5 The field below the hyperbola (slash field) will express the condition that hydrogen mass flow rates ran short, and if it goes into the starting field, it will open a purge valve 11.

[0040] The flow chart of drawing 6 shows the above-mentioned purge control flow, and measures the propagation times t1 and t2 of a supersonic wave at step S1 first. and the step S -- a volumetric flow rate Q is calculated based on said propagation times t1 and t2, and hydrogen gas concentration x is calculated by step S4-6 by the concurrency 2 and 3.

[0041] At step S2, mean velocity V is calculated based on said propagation times t1 and t2 (formula (3)), and a volumetric flow rate Q is calculated based on this mean velocity V (formula (5)). At step S3, amendment according to a pressure and temperature is performed to said volumetric flow rate Q, and it asks for volumetric flow rate Q'. In step S4, acoustic velocity C is calculated based on said propagation times t1 and t2 (formula (4)).

[0042] Said acoustic velocity C is changed into mean density rhoav at step S5 (refer to drawing 3 ). At step S6 (concentration presumption means), the hydrogen gas concentration x (and impurity gas concentration y) is calculated based on said mean density rhoav (formula (10)). At step S7 (hydrogen mass flow rate presumption means), a hydrogen gas mass flow rate is calculated based on said volumetric flow rate Q', and the hydrogen gas concentration x and the hydrogen gas consistency rho x.

[0043] At step S8 (threshold setting means), the threshold of said hydrogen gas mass flow rate is set up according to the generation-of-electrical-energy current at that time. It distinguishes whether the hydrogen gas mass flow rate calculated at step S7 is below the threshold set up at step S8, and with [ a hydrogen gas mass flow rate ] a threshold [ below ], it is made to purge in step S9 by progressing to step S10 (purge control means), and opening a purge valve 11 wide.

[0044] By the way, although an unnecessary purge is avoidable by making it purge as mentioned above in the state of steady operation when a hydrogen gas mass flow rate becomes below a threshold, the load effect of a fuel cell is especially large in a vehicle use fuel cell system, and there is much fluctuation frequency. And at the time of a load effect, it will be based on the value of the threshold set up corresponding to the generation-of-electrical-energy current of a fuel cell, and if it becomes easy to enter into the condition that hydrogen mass flow rates ran short, a load is interlocked with and a purge valve 11 is opened wide frequently, the hydrogen fuel gas discharged by purge valve disconnection will also increase unnecessarily.

[0045] Even if it is in the above-mentioned hydrogen mass flow rate insufficient field temporarily in the condition with little impurity gas abundance in the circulatory system to lack of the above transitional hydrogen mass flow rates, it will be good to make it not open a purge valve. Said impurity gas abundance can presume the amount corresponding to impurity gas abundance to be impurity gas concentration y shown by the formula (11) from a pressure p and temperature T, if it considers that the volume V which consists of ducts of a reflux system etc. is fixed (impurity gas abundance presumption means).

[0046] Here, the above-mentioned impurity gas abundance is not a flow rate (L/min), and it will not correspond with the impurity gas concentration y itself, and the value which amended impurity gas concentration y at a pressure p and temperature T will correspond to abundance.

[0047]

[Equation 1]

「不純物ガス存在量」 $\propto y \cdot (p/p_0) \cdot (T_0/T) \cdot V \dots\dots (12)$

[0048] If it is made to perform a purge when the value corresponding to the impurity gas abundance calculated as mentioned above when a hydrogen mass flow rate becomes below a threshold is beyond a threshold, it will be avoided that a purge is unnecessarily performed at the time of a load effect. In addition, when a pressure and temperature are assumed to be fixed to the purge field shown in drawing 5 in the purge control only based on a hydrogen mass flow rate, since it is impurity gas concentration y=1-hydrogen gas concentration, by the purge based on an above-mentioned hydrogen mass flow rate and impurity gas abundance, a purge will be performed in the field limited rather than it was shown in the slash field surrounded by b which is equivalent to the threshold of impurity gas concentration like drawing 7 .

[0049] The flow chart of drawing 8 shows the purge control according to an above-mentioned hydrogen mass flow rate and impurity gas abundance, and processing of steps S21-S29 is performed like step S1 of

the flow chart of said drawing 6 - S9. At step S29 (impurity gas abundance presumption means), if it is distinguished that a hydrogen mass flow rate is below a threshold, it will progress to step S30 and impurity gas abundance will be calculated.

[0050] It distinguishes whether it is beyond the threshold said impurity gas abundance was beforehand remembered to be, and with [ said impurity gas abundance ] a threshold [ beyond ], it is made to purge by progressing to step S32 at step S31. By the way, although considered as the configuration which makes the flow rate of the mixed gas of hydrogen and impurity gas measure by the downstream of a dehumidifier 8 by the system configuration shown in drawing 1 when the mixed gas of the circulatory system is not dehumidified but it has become three components of hydrogen, a steam, and impurity gas As shown in drawing 9 or drawing 10, even when detecting a quantity of gas flow in the upstream location of a dehumidifier 8, it can ask for mean density rhoav to the hydrogen gas concentration x and impurity gas concentration y which are called for by making steam concentration z into known based on said propagation times t1 and t2.

[0051] With the operation gestalt shown in drawing 9, when the hygrometer 35 is made to have infix near the upstream of a flowmeter 30 and it has this hygrometer 35 (humidity detection means), it can ask for the steam concentration z from the detection result of this humidity, and a pressure p. Moreover, with the operation gestalt shown in drawing 10, although it does not have a hygrometer 35, when the inside of the circulatory system is in a saturated steam condition, it can ask for the saturated water vapor pressure in temperature T based on a pressure p and temperature T, and a steam partial pressure (concentration) can be presumed.

[0052] Here, since the propagation time of the supersonic wave in hydrogen, impurity gas, and the mixed gas of a steam will be measured, mean density rhoav based on the propagation times t1 and t2 is mean density rhoav of 3 component mixed gas, and a flowmeter 30 will be changed as follows, if said formula (8) and (9) set the consistency of a steam to rhoz.

$$x+y+z=1 \dots (13)$$

$$X\text{-rho } x+y-\text{rho } y+z-\text{rho } z=\text{rho av} \dots (14)$$

$$\text{Here, an upper type is transformed and it is } x+y=1-z. \dots (15)$$

$$X\text{-rho } x+y-\text{rho } y+z-\text{rho } z=\text{rho av}-z-\text{rho } z \dots (16)$$

$$\text{When it carries out and this is solved about Unknown x and y, it is. } x=[(1-z) \text{ rho } y+z-\text{rho } z-\text{rho av}] /(\text{rho } y-\text{rho } x) \dots (17)$$

$$y=[\text{rho av}-z-\text{rho } z-(1-z) \text{ rho } y] /(\text{rho } y-\text{rho } x) \dots (18)$$

If a next door and the steam concentration z are known, the hydrogen gas concentration x and impurity gas concentration y will be called for from an upper type.

[0053] Drawing 11 makes steam concentration z known in the configuration whose ultrasonic flowmeter 30 measures the propagation time in hydrogen, impurity gas, and the mixed gas of a steam, and the purge control which calculates the hydrogen concentration x from this steam concentration z is shown. If mean density rhoav is calculated at step S25, the flow chart shown in drawing 11 will calculate the steam concentration z from a pressure and temperature by the following step S26A (concentration presumption means) based on a hygrometer 35, when the inside of the circulatory system is in a saturated steam condition.

[0054] And in step S26B, the hydrogen gas concentration x is calculated using said steam concentration z. At each step other than above-mentioned step S26A and step S26B, the same processing as drawing 8 is performed. In addition, when the humidity within the circulatory system, a pressure, and temperature are abbreviation regularity, even if it does not form a hygrometer 35, a pressure gage 36, and a thermometer 37, it is possible to consider beforehand that the steam concentration z is constant value, and to ask for the hydrogen gas concentration x and impurity gas concentration y.

[0055] Moreover, it is good also as a configuration which makes a flowmeter 30 infix in the supply line 5 which constitutes the hydrogen gas circulatory system as shown in drawing 12. In this case, if piling of the threshold of the hydrogen mass flow rate set up according to a generation-of-electrical-energy current is carried out a supplied part from a supply line 3, it can make purge control perform like said operation gestalt, since the hydrogen mass flow rate measured becomes the part which flows back with an ejector 4, and the supplied sum total from the supply line 3 corresponding to a consumed part in the fuel cell stack 20.

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[Translation done.]

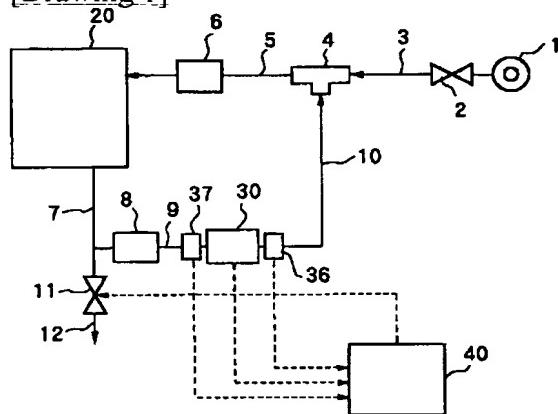
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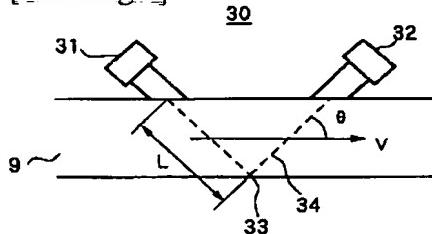
1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. \*\*\* shows the word which can not be translated.
3. In the drawings, any words are not translated.

## DRAWINGS

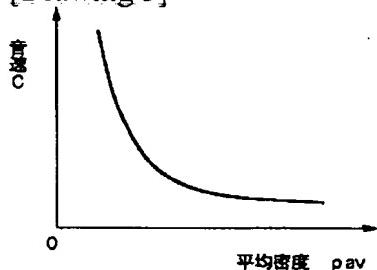
[Drawing 1]



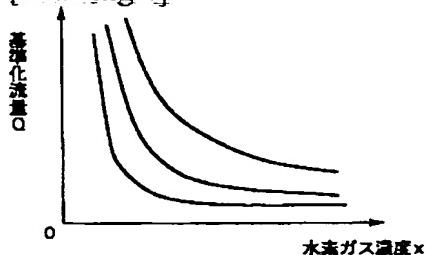
[Drawing 2]



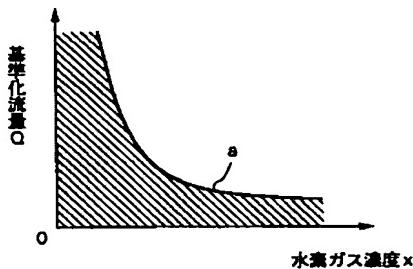
[Drawing 3]



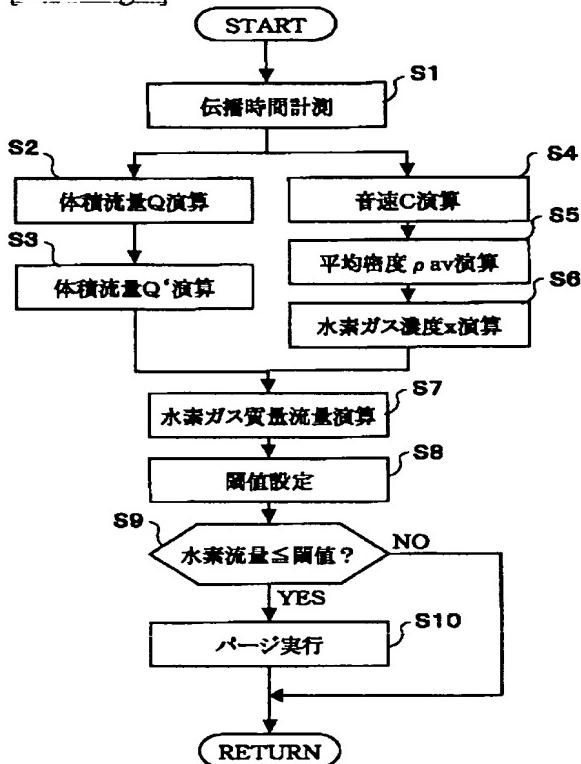
[Drawing 4]



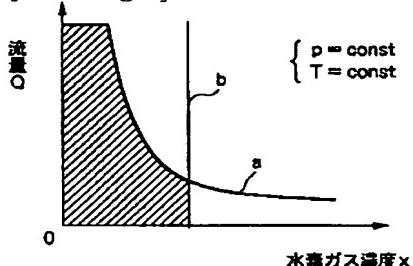
[Drawing 5]



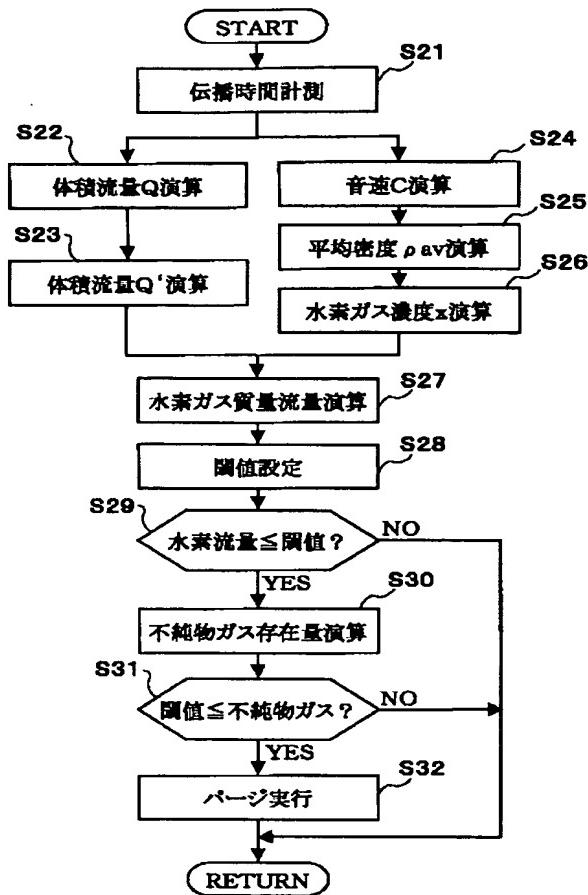
[Drawing 6]



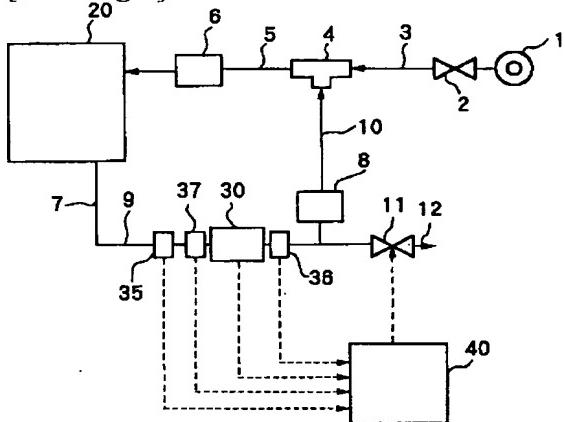
[Drawing 7]



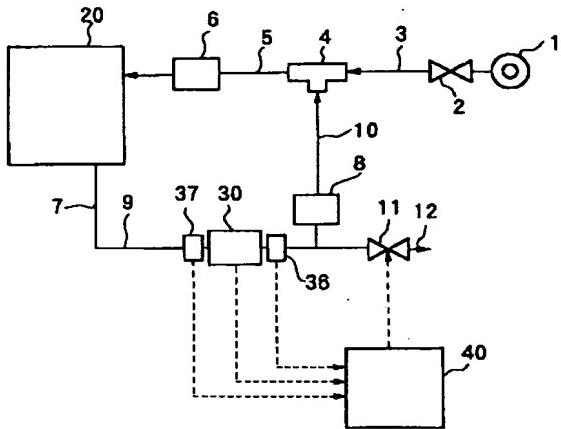
[Drawing 8]



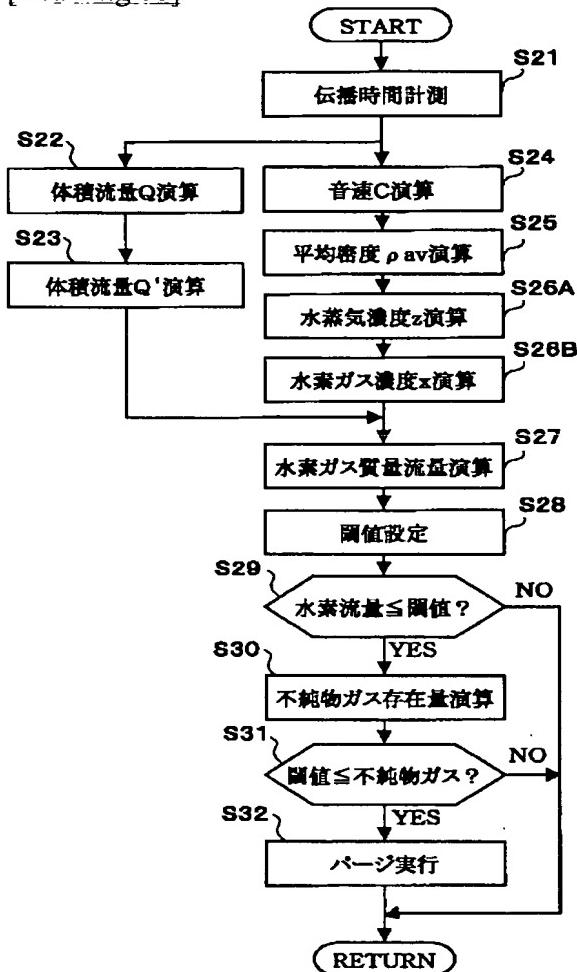
[Drawing 9]



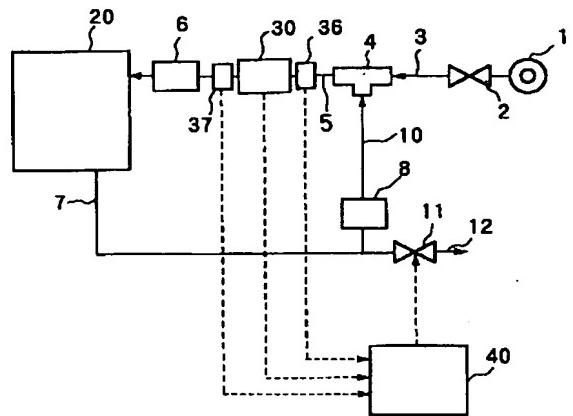
[Drawing 10]



[Drawing 11]



[Drawing 12]



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[Translation done.]

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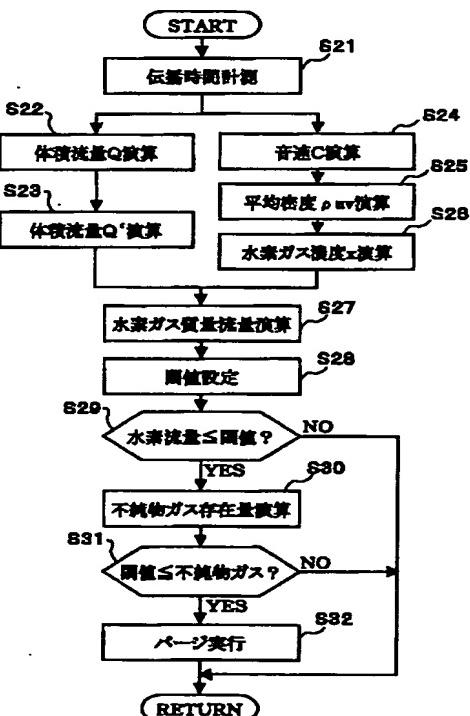
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(54)【発明の名称】 燃料電池システム及び制御方法

(57)【要約】

【課題】 燃料電池システムにおいて、バージによる燃料ガスの無用な損失を低減させて燃料電池システムのエネルギー効率を高めることを目的とする。

【解決手段】 超音波流量計で計測される混合ガス中の超音波の伝播時間に基づいて体積流量を演算すると共に、混合ガスの平均密度を求める。そして、前記平均密度から水素ガス濃度及び不純物ガス濃度を演算すると共に、該水素ガス濃度と体積流量から水素ガスの質量流量を演算し、不純物ガス濃度、圧力、温度から不純物ガス存在量を演算する。ここで、水素質量流量が閾値以下で、かつ、不純物ガス存在量が閾値以上であるときに、バージを行わせる。



1

(2)

2

## 【特許請求の範囲】

【請求項 1】燃料電池において発電に使用されなかった水素を、前記燃料電池へ水素を供給する通路に還流させる水素還流経路を備えた燃料電池システムであって、前記水素還流経路に設けられるガスバージ弁と、前記水素還流経路を含む水素循環系内における水素質量流量を推定する水素質量流量推定手段と、前記水素質量流量が閾値以下になったときに、前記ガスバージ弁を開放するバージ制御手段と、を含んで構成されたことを特徴とする燃料電池システム。

【請求項 2】前記水素循環系内の混合ガス中の不純物ガス存在量を推定する不純物ガス存在量推定手段を備え、前記バージ制御手段が、前記水素質量流量が閾値以下で、かつ、前記不純物ガス存在量が閾値以上になったとき、前記ガスバージ弁を開放することを特徴とする請求項 1 記載の燃料電池システム。

【請求項 3】前記水素質量流量の閾値を、前記燃料電池の運転条件に応じて設定する閾値設定手段を設けたことを特徴とする請求項 1 又は 2 記載の燃料電池システム。

【請求項 4】前記閾値設定手段が、前記運転条件としての発電電流に応じて前記水素質量流量の閾値を設定することを特徴とする請求項 3 記載の燃料電池システム。

【請求項 5】前記水素質量流量推定手段及び／又は不純物ガス存在量推定手段が、前記水素循環系内の混合ガス中の音速を求めると共に、該音速に基づいて前記混合ガスの平均密度を求め、該平均密度に基づいて前記混合ガス中の水素濃度及び／又は不純物ガス濃度を推定する濃度推定手段を含んで構成され、

該濃度推定手段によって求められる濃度に基づいて水素質量流量及び／又は不純物ガス存在量を推定することを特徴とする請求項 1～4 のいずれか 1 つに記載の燃料電池システム。

【請求項 6】前記水素質量流量推定手段が、前記水素循環系内の混合ガス中における超音波の伝播時間に基づいて前記混合ガスの体積流量を検出する体積流量検出手段を含んで構成され、

前記濃度推定手段が、前記体積流量検出手段による伝播時間の計測結果に基づいて音速を求めて、前記水素濃度及び／又は不純物ガス濃度を推定することを特徴とする請求項 5 記載の燃料電池システム。

【請求項 7】前記水素循環系内の混合ガス中における超音波の伝播時間に基づいて前記混合ガスの体積流量を検出する体積流量検出手段と、

前記混合ガスの圧力を検出する圧力検出手段と、前記混合ガスの温度を検出する温度検出手段と、

前記超音波の伝播時間に基づいて前記混合ガス中の音速を求めると共に、該音速に基づいて前記混合ガスの平均密度を求め、該平均密度に基づいて前記混合ガス中の水

素ガス濃度及び不純物ガス濃度を推定する濃度推定手段と、を備え、

前記水素質量流量推定手段が、前記体積流量、圧力、温度及び水素ガス濃度に基づいて水素質量流量を推定し、前記不純物ガス存在量推定手段が、前記圧力、温度及び不純物ガス濃度に基づいて前記不純物ガス存在量を推定することを特徴とする請求項 2 記載の燃料電池システム。

【請求項 8】前記水素還流経路に除湿手段を介装し、前記濃度推定手段が、前記除湿手段の下流側における超音波の伝播時間に基づいて水素濃度及び／又は不純物ガス濃度を推定することを特徴とする請求項 5～7 のいずれか 1 つに記載の燃料電池システム。

【請求項 9】前記超音波の伝播時間の計測部位近傍に、前記混合ガスの湿度を検出する湿度検出手段を設け、前記濃度推定手段が、前記湿度に基づいて求められる水蒸気濃度と前記平均密度とに基づいて、前記水素ガス濃度及び／又は不純物ガス濃度を推定することを特徴とする請求項 5～7 のいずれか 1 つに記載の燃料電池システム。

【請求項 10】前記超音波の伝播時間の計測部位近傍に、前記混合ガスの圧力を検出する圧力検出手段及び混合ガスの温度を検出する温度検出手段を設け、前記濃度推定手段が、飽和水蒸気状態での水蒸気濃度を前記圧力及び温度から求め、該水蒸気濃度と前記平均密度とに基づいて、前記水素ガス濃度及び／又は不純物ガス濃度を推定することを特徴とする請求項 5～7 のいずれか 1 つに記載の燃料電池システム。

【請求項 11】前記濃度推定手段が、前記混合ガスの水蒸気濃度を予め定められた一定値とし、該水蒸気濃度と前記平均密度とに基づいて、前記水素ガス濃度及び／又は不純物ガス濃度を推定することを特徴とする請求項 5～7 のいずれか 1 つに記載の燃料電池システム。

【請求項 12】燃料電池において発電に使用されなかった水素を、前記燃料電池へ水素を供給する通路に還流させる水素還流経路を備えた燃料電池システムにおいて、前記水素還流経路を含む水素循環系内における水素質量流量を推定し、

前記水素質量流量が閾値以下になったときに、前記水素還流経路から混合ガスをバージさせることを特徴とする燃料電池システムの制御方法。

【請求項 13】燃料電池において発電に使用されなかった水素を、前記燃料電池へ水素を供給する通路に還流させる水素還流経路を備えた燃料電池システムにおいて、前記水素還流経路を含む水素循環系内における水素質量流量を推定し、

前記水素循環系内の混合ガス中の不純物ガス存在量を推定し、

前記水素質量流量が閾値以下で、かつ、前記不純物ガス

50

存在量が閾値以上になったとき、前記水素還流経路から混合ガスをバージさせることを特徴とする燃料電池システムの制御方法。

【請求項14】前記水素質量流量の閾値を、前記燃料電池の発電電流に応じて設定することを特徴とする請求項12又は13記載の燃料電池システムの制御方法。

【請求項15】前記水素循環系内の混合ガス中における超音波の伝播時間に基づいて、前記混合ガス中の音速を求め、

該音速に基づいて前記混合ガスの平均密度を求め、  
該平均密度に基づいて前記混合ガス中の水素濃度及び／又は不純物ガス濃度を推定し、

前記濃度に基づいて水素質量流量及び／又は不純物ガス存在量を推定することを特徴とする請求項12～14のいずれか1つに記載の燃料電池システムの制御方法。

【発明の詳細な説明】

【0001】

【発明の属する技術分野】本発明は、燃料電池システム及び制御方法に関し、詳しくは、燃料ガスを循環して燃料電池に還流させて再利用するタイプの燃料電池システムにおいて、還流系からのバージを制御する技術に関する。

【0002】

【従来の技術】従来から、燃料電池システムにおいて、燃料電池で消費されなかった水素を、燃料電池に還流して再利用を図ることが行われている（特開平10-055814号公報及び特開2000-58092号公報参照）。また、上記のように燃料ガスを還流させて再利用するシステムの場合、燃料ガスを循環し続けるにしたがって循環ライン内に不純物ガス（例えば燃料電池の発電作用には不必要的窒素ガス）が徐々に蓄積・増加していくことが知られている。

【0003】このような不必要的ガスを循環させ続けることは循環動力を無駄に使うばかりでなく、燃料電池に供給される燃料中に占める不純物ガスの割合が増加することで燃料電池の出力低下を招くことになる。そこで、前記特開2000-058092号に開示される構成では、燃料ガスの循環ラインから不純物ガスを含む燃料ガスをバージさせるバージ弁を備え、循環ライン中の不純物ガス濃度が一定値を超えたときに前記バージ弁を開放させるようになっていた。

【0004】

【発明が解決しようとする課題】ところで、燃料ガスを還流させて再利用する燃料電池システムにおいては、燃料ガス供給源からの水素供給流量をQ(L/min)とすると、このQ(L/min)に対してある比率 $\alpha$ の流量 $\alpha Q(L/min)$ がエジェクタによって吸込まれて還流され、燃料電池には合計流量である $(1+\alpha)Q(L/min)$ が流入する。

【0005】そして、燃料電池にてQ(L/min)の

水素が消費され、消費されずに残った $\alpha Q(L/min)$ が未利用ガスとして燃料電池から排出されて循環するという関係にある。すなわち、燃料ガス供給源から流入する水素流量Q(L/min)が燃料電池で消費され、この流量Q(L/min)に依存した量 $\alpha Q(L/min)$ が循環している。

【0006】一方、特開2000-012059号公報に開示されるように、燃料電池に流入する水素質量流量が燃料電池の発電効率に関係しており、燃料電池で消費される水素質量流量以上の量を燃料電池に流入することが求められ、また、燃料電池の急激な負荷増大に対応するための余裕分も必要なことから、循環される水素質量流量が所定以上であることが要求される。

【0007】換言すれば、循環される水素質量流量（余裕分）が所定以上であれば、燃料電池の発電に影響がないため、循環ラインからのバージは必要ないことになるが、従来のように、不純物ガス濃度に基づいてバージを制御する構成では、不純物ガス濃度が同じでも燃料電池における消費量によって循環ラインにおける水素質量流量が変化することに対応できないため、適切なタイミングでバージを行わせることができないという問題があった。

【0008】また、従来のように不純物ガス濃度に基づいてバージを制御する構成では、過渡的な不純物ガス濃度の変化に影響されてバージが行われ、無用な燃料ガスの損失を増大させる可能性があった。例えば、循環ラインの不純物ガスがある量蓄積されている状態で、燃料電池の負荷の増大変化があると、一時的に循環ラインの水素質量が低下し、それによって相対的に不純物ガス濃度が増大するため、従来制御では、係る一時的な不純物ガス濃度の増大に基づいて、バージが行われる可能性がある。

【0009】しかし、有効なバージを実行するという観点からすると、不純物ガスの蓄積存在量が所定量以下の場合はバージを控えることが望まれるため、従来制御では、頻繁なバージ制御によって無用な燃料ガスの損失を増大させる可能性があった。本発明は上記問題点に鑑みされたものであり、燃料ガスの無用な損失を低減させて燃料電池システムのエネルギー効率を高めることが可能な燃料電池システム及び制御方法を提供することを目的とする。

【0010】

【課題を解決するための手段】上記目的を達成するため、請求項1、12記載の発明では、水素還流経路を含む水素循環系内における水素質量流量を推定し、前記水素質量流量が閾値以下になったときに、水素還流経路から混合ガスをバージさせる構成とした。上記構成によると、水素循環系内の混合ガス（水素ガス、不純物ガス、水蒸気）の流量変化に応じて変化する水素質量流量が閾値以下となり、発電効率を維持できなくなると、バージ

を行って燃料ガスと共に不純物ガスをバージさせる。

【0011】請求項2、13記載の発明では、水素循環系内における水素質量流量及び不純物ガス存在量をそれ推定し、前記水素質量流量が閾値以下で、かつ、前記不純物ガス存在量が閾値以上になったときに、水素還流経路から混合ガスをバージさせる構成とした。上記構成によると、たとえ燃料電池の負荷変化等によって水素質量流量が閾値以下になっても、不純物ガス存在量が閾値を下回るときには、バージが行われない。

【0012】請求項3記載の発明では、前記水素質量流量の閾値を、前記燃料電池の運転条件に応じて設定する構成とした。上記構成によると、必要とされる水素質量流量が、燃料電池の運転条件に応じて変化することに対応させるべく、バージを行わせる水素質量流量の閾値を変化させる。

【0013】請求項4、14記載の発明では、燃料電池の発電電流に応じて前記水素質量流量の閾値を設定する構成とした。上記構成によると、発電電流に応じて発電効率の維持のために必要とされる水素質量流量が変化することに対応して、水素質量流量の閾値が変更される。請求項5、15記載の発明では、水素質量流量及び／又は不純物ガス存在量の推定に用いる水素濃度及び／又は不純物ガス濃度を、水素循環系内の混合ガス中の音速に基づいて求められる混合ガスの平均密度に基づいて推定させる構成とした。

【0014】上記構成によると、混合ガス中における音速から混合ガスの平均密度が分かり、例えば、混合ガスが水素と不純物ガスとからなる場合には、水素濃度と不純物ガス濃度との合計が1で、各ガスの濃度と密度との乗算値を加算した結果が平均密度となるから、これらの式に基づいて水素濃度及び不純物ガス濃度を求めることができる。

【0015】請求項6記載の発明では、水素質量流量の推定に、水素循環系内の混合ガス中における超音波の伝播時間に基づいて体積流量を検出する手段を用いる構成とし、濃度推定に前記体積流量の検出のための伝播時間の計測結果を用いる構成とした。上記構成によると、体積流量の検出のために計測される伝播時間に基づいて、同時に、水素濃度及び／又は不純物ガス濃度の推定が行われる。

【0016】請求項7記載の発明では、超音波の伝播時間に基づいて体積流量を検出する手段と、混合ガスの圧力を検出する手段と、混合ガスの温度を検出する手段と、を備え、前記伝播時間に基づいて混合ガス中の音速を求め、該音速に基づいて求められる平均密度に基づいて水素ガス濃度及び不純物ガス濃度を推定し、前記体積流量、圧力、温度及び水素ガス濃度に基づいて水素質量流量を推定すると共に、前記圧力、温度及び不純物ガス濃度に基づいて不純物ガス存在量を推定する構成とした。

【0017】上記構成によると、体積流量の検出に伴って濃度推定が行われ、また、圧力、温度の変化に対応して、水素質量流量及び不純物ガス存在量が推定される。請求項8記載の発明では、除湿手段の下流側での伝播時間に基づいて、水素濃度及び／又は不純物ガス濃度を推定させる構成とした。上記構成によると、水素循環系の混合ガスが、水素、不純物ガス、水蒸気からなる場合に、伝播時間の計測部位の上流側で除湿されることで、混合ガスが水素と不純物ガスとの2成分から構成されることになり、伝播時間から水素の密度と不純物ガスの密度との平均が求められることになる。従って、未知数が水素濃度及び不純物ガス濃度のみとなり、濃度の方程式と密度の方程式とから、水素濃度及び不純物ガス濃度が求められることになる。

【0018】請求項9記載の発明では、伝播時間の計測部位近傍で湿度を検出し、該湿度に基づいて求められる水蒸気濃度と伝播時間から求められる平均密度とに基づいて、水素ガス濃度及び／又は不純物ガス濃度を推定する構成とした。上記構成によると、平均密度が、水素、不純物ガス、水蒸気の平均密度として求められる構成であっても、水蒸気の濃度を既知とすることで、水素濃度及び不純物ガス濃度が求められることになる。

【0019】請求項10記載の発明では、伝播時間の計測部位近傍で、圧力及び温度を検出し、飽和水蒸気状態での水蒸気濃度を前記圧力及び温度から求め、該水蒸気濃度と伝播時間から求められる平均密度とに基づいて、水素ガス濃度及び／又は不純物ガス濃度を推定する構成とした。上記構成によると、水素循環系内が略飽和水蒸気状態であれば、圧力及び温度から水蒸気濃度が求められ、水蒸気の濃度を既知とすることで、水素濃度及び不純物ガス濃度が求められることになる。

【0020】請求項11記載の発明では、混合ガスの水蒸気濃度を予め定められた一定値とし、該水蒸気濃度と伝播時間から求められる平均密度とに基づいて、水素ガス濃度及び／又は不純物ガス濃度を推定する構成とした。上記構成によると、水蒸気の濃度を一定値と仮定することで、水素濃度及び不純物ガス濃度が求められる。

#### 【0021】

【発明の効果】請求項1、12記載の発明によると、水素循環系における水素質量流量に基づいてバージを制御するので、発電効率を維持できる水素質量流量が確保されている状態でバージが行われることを回避でき、以って、無用なバージによる水素ガスの損失を低減させることができるという効果がある。

【0022】請求項2、13記載の発明によると、水素質量流量が低下しても、不純物ガス存在量が少ない場合にはバージが行われないので、過渡的に水素質量流量が低下したときに、バージすべき不純物ガス存在量が少ないのにバージが行われてしまうことを回避でき、有効なバージのみを行わせて、より一層無用なバージによる水

素ガスの損失を低減させることができるという効果がある。

【0023】請求項3、4、14記載の発明によると、発電効率を維持でき、また、負荷変動に対応できるだけの余裕分を確保できる水素質量流量になっているか否かに基づいて、バージを行わせるので、燃料電池の運転条件(負荷)の変化に応じて常に最適なタイミングでバージを行わせることができるという効果がある。請求項5、15記載の発明によると、混合ガス中の音速から水素濃度及び/又は不純物ガス濃度を推定するので、水素質量流量・不純物ガス存在量の推定に必要な濃度情報を音速に基づき得ることができるという効果がある。

【0024】請求項6記載の発明によると、水素質量流量を推定するための体積流量の検出に伴って同時に濃度情報を得ることができ、簡単な構成で水素質量流量・不純物ガス存在量の推定を行わせることができるという効果がある。請求項7記載の発明によると、水素質量流量を推定するための体積流量の検出に伴って同時に濃度情報を得ることができ、かつ、水素循環系内の圧力、温度の変化に対応して水素質量流量及び不純物ガス存在量を高精度に推定することができるという効果がある。

【0025】請求項8記載の発明によると、混合ガス中の超音波の伝播時間に基づき水素濃度及び/又は不純物ガス濃度を推定させる構成において、除湿後の混合ガスにおける伝播時間を計測させて、水蒸気ガスの濃度測定を行うことなく、水素濃度及び不純物ガス濃度を推定することができるという効果がある。請求項9記載の発明によると、湿度の検出結果から水蒸気濃度を求ることで、水蒸気を含む混合ガス中における超音波の伝播時間から水素濃度・不純物ガス濃度を推定することができるという効果がある。

【0026】請求項10記載の発明によると、水素循環系内が飽和水蒸気状態であるときに、圧力、温度から簡単に水蒸気濃度を求めて、水蒸気を含む混合ガス中における超音波の伝播時間から水素濃度・不純物ガス濃度を推定することができるという効果がある。請求項11記載の発明によると、水素循環系内の湿度、圧力、温度が略一定と見なせる条件下であるときに、水蒸気を含む混合ガス中における超音波の伝播時間から水素濃度・不純物ガス濃度を簡単に推定させることができるという効果がある。

### 【0027】

【発明の実施の形態】以下に本発明の実施の形態を図に\*

$$V = L / (2 \cos \theta) * (1/t_1 - 1/t_2) \quad \dots \dots (3)$$

$$C = L / 2 * (1/t_1 + 1/t_2) \quad \dots \dots (4)$$

となる。

【0032】従って、水素ガス還流路9内における平均体積流量Q(m<sup>3</sup>/s)は、管路断面積をS、流量補正係数をKとすると、

$$Q = V \cdot S \cdot K \quad \dots \dots (5)$$

\*基づいて説明する。図1は、実施形態における燃料電池システムを示す図である。この図1において、水素ガス供給源1からの水素ガスは、供給調圧バルブ2、水素ガス供給ライン3、エジェクタ4、水素ガス供給ライン5を介して、燃料電池スタック20の燃料極(図示省略)に導入され、燃料電池スタック20にて消費されなかった燃料ガスは、水素ガス還流路7、9、10(水素還流経路)を介し、前記エジェクタ4によって水素ガス供給ライン5に還流される。

【0028】上記構成によって、燃料電池スタック20において発電に使用されなかった水素は、再び燃料電池20に還流され、水素の循環系が水素ガス還流路7、9、10及び水素ガス供給ライン5によって構成される。前記水素ガス還流路7から分岐延設される排出ライン12には、バージ弁11が介装されている。

【0029】前記排出ライン12の分岐部よりも下流側の水素ガス還流路9には、上流側から順に、除湿器8(除湿手段)、温度計37(温度検出手段)、流量計30(体積流量検出手段)、圧力計36(圧力検出手段)が介装されている。また、前記水素ガス供給ライン5には、加湿器6が介装されている。信号演算処理部40は、前記温度計37、流量計30、圧力計36からの検出信号を入力し、後述する演算処理に基づいて前記バージ弁11の開放を制御する。

【0030】図2は、前記流量計30の詳細を示す。前記流量計30は、本実施形態において超音波式流量計であり、超音波受発信器31、32を備え、上流側の超音波受発信器31から下流側に向けて斜めに発した音波が、超音波反射部33で反射して下流側の超音波受発信器32から上流側に向けて斜めに発した音波が、超音波反射部33で反射して上流側の超音波受発信器31に受信されるようになっており、下流側に向かう超音波の伝播時間t1と、上流側に向かう超音波の伝播時間t2とが計測されるようになっている。

【0031】ここで、水素ガス還流路9の軸線と超音波との角度をθ、ガスの音速をC、超音波の伝播経路34の長さをL、水素ガス還流路9での平均流速をVとすると、前記伝播時間t1、t2(sec)は、

$$t_1 = L / (C + V \cos \theta) \quad \dots \dots (1)$$

$$t_2 = L / (C - V \cos \theta) \quad \dots \dots (2)$$

で表され、上記の2式から、平均流速V(m/s)、音速C(m/s)を求める

$$V = L / (2 \cos \theta) * (1/t_1 - 1/t_2) \quad \dots \dots (3)$$

$$C = L / 2 * (1/t_1 + 1/t_2) \quad \dots \dots (4)$$

で求められるが、前記平均体積流量Qは、基準温度及び基準圧力状態での体積流量であるため、そのときの圧力p及び温度Tに応じた補正を施して、Q'(ノルマル立方メートル/秒)を算出する。

【0033】

$$Q' (\text{Nm}^3/\text{s}) = Q \times (p/p_0) \times (T_0/T) \quad \dots \dots (6)$$

尚、上式で、 $p_0$ は基準圧力を、 $T_0$ は基準温度を示す。本実施形態では、水素質量流量  $dmH_2/dt$ に基づいてバージを制御する構成となっており、水素ガス濃度を  $x$ 、水素密度を  $\rho_x$  とすると、水素質量流量  $dmH_2/dt$  ( $\text{kg}/\text{s}$ ) は、

$$dmH_2/dt = x \cdot \rho_x \cdot Q' \quad \dots \dots (7)$$

として算出される（水素質量流量推定手段）。

【0034】ここで、循環系には、水素、水蒸気、不純物ガス（窒素）からなる混合ガスが流れるが、前記超音波式の流量計30は除湿器8の下流側に配設されるから、前記音速Cは、水蒸気を除く水素と不純物ガス（窒素）との2成分からなる混合ガス中における音速Cとなる。また、音速Cを求めた混合ガスの成分が既知である場合には、音速Cは、各成分の混合比（濃度）の関数で表され（特開2000-304732号参照）、水素と不純物ガス（窒素）の2成分の場合、係る混合ガスの平均密度  $\rho_{av}$  と音速Cとは、図3に示すような相関になる。

【0035】従って、混合ガスでの音速Cが分かれれば、図3に従って混合ガスの平均密度  $\rho_{av}$  が求まり、平均密度  $\rho_{av}$  から各成分の濃度を求めることができる。即ち、水素ガスの濃度を  $x$ 、水素ガスの密度を  $\rho_x$  とし、不純物ガスの濃度を  $y$ 、不純物ガスの密度を  $\rho_y$  とすると、混合ガスは水素ガスと不純物ガスとの2成分からなるから、

$$x + y = 1 \quad \dots \dots (8)$$

$$x \cdot \rho_x + y \cdot \rho_y = \rho_{av} \quad \dots \dots (9)$$

であるから、

$$x = (\rho_y - \rho_{av}) / (\rho_y - \rho_x) \quad \dots \dots (10)$$

$$y = (\rho_{av} - \rho_x) / (\rho_y - \rho_x) \quad \dots \dots (11)$$

となり、水素ガスの密度  $\rho_x$  及び不純物ガスの密度  $\rho_y$  と、音速Cから求められる平均密度  $\rho_{av}$  とから、水素ガス濃度  $x$  及び不純物ガス濃度  $y$  を算出することができる（濃度推定手段）。

【0036】尚、上記水素ガスの濃度  $x$  及び不純物ガス濃度  $y$  を求めるときに用いる水素ガスの密度  $\rho_x$  及び不純物ガスの密度  $\rho_y$  は、そのときの圧力・温度から設定される。上記のようにして水素ガス濃度  $x$  及び不純物ガス濃度  $y$  が求められると、係る濃度  $x$ 、 $y$  に前記体積流量  $Q'$  を乗算することで、水素ガス体積流量  $Q_x'$  及び不純物ガス体積流量  $Q_y'$  が求められ、該体積流量  $Q_x'$ 、 $Q_y'$  にガス密度  $\rho_x$ 、 $\rho_y$  を乗算することで、水素質量流量と不純物ガス質量流量が求められる。

【0037】燃料電池に供給する水素質量流量は燃料電池の発電効率に影響を与えるため、燃料電池で消費される水素質量流量以上の量を燃料電池に流入させることが求められ、また、燃料電池の急激な負荷増大に対応するための余裕分も必要なことから、循環ラインにおける水素質量流量、即ち、消費量分に付加される余裕分が所定

以上であることが要求される。

【0038】一方、還流ガス流量は、エジェクタ4によってある比率の流量が自動的に還流されるようになっているため、還流ガスの中の水素質量流量が上記要求に見合った量よりも少ないと、燃料電池では、必要な水素ガス量が不足し、逆に、不必要的ガス成分（不純物ガス）が充満することになる。そこで、前記信号演算処理部40では、上記のようにして算出した水素質量流量と閾値とを比較し、水素質量流量が閾値以下になると、燃料電池で必要な水素量が確保できる状態となるまで（例えば一定時間）、バージ弁11を開いて循環系内に溜まった不純物ガスをバージする（バージ制御手段）。

【0039】ここで、燃料電池にとって必要な水素ガス質量流量とは、燃料電池の運転状態に依存しており、燃料電池を変換器と見なすと、その出力である発電電流値に対応した量に設定するのが合理的であり、前記閾値を発電電流に応じて変更すると良い（閾値設定手段）。図4に示すように、前記水素ガス濃度  $x$  と体積流量  $Q'$  とを変数とすると、水素質量流量一定の状態は双曲線群で表され、ある発電電流値に対応した水素質量流量  $a$  で定常運転する場合を想定すると、図5に示すように、その双曲線以下の領域（斜線領域）は水素質量流量が不足した状態を表すことになり、係る領域に入るとバージ弁11を開くようとする。

【0040】図6のフローチャートは、上記バージ制御の流れを示すものであり、まず、ステップS1では、超音波の伝播時間  $t_1$ 、 $t_2$  の計測を行う。そして、ステップS2、3では、前記伝播時間  $t_1$ 、 $t_2$  に基づき体積流量  $Q$  の演算を行い、同時に並行で、ステップS4～6では、水素ガス濃度  $x$  の演算を行う。

【0041】ステップS2では、前記伝播時間  $t_1$ 、 $t_2$  に基づき平均流速  $V$  を演算し（式（3））、該平均流速  $V$  に基づいて体積流量  $Q$  を演算する（式（5））。ステップS3では、前記体積流量  $Q$  に圧力、温度に応じた補正を施して、体積流量  $Q'$  を求める。ステップS4では、前記伝播時間  $t_1$ 、 $t_2$  に基づき音速Cを演算する（式（4））。

【0042】ステップS5では、前記音速Cを平均密度  $\rho_{av}$  に変換する（図3参照）。ステップS6（濃度推定手段）では、前記平均密度  $\rho_{av}$  に基づいて水素ガス濃度  $x$ （及び不純物ガス濃度  $y$ ）を演算する（式（10））。ステップS7（水素質量流量推定手段）では、前記体積流量  $Q'$ 、水素ガス濃度  $x$  及び水素ガス密度  $\rho_x$  に基づいて水素ガス質量流量を演算する。

【0043】ステップS8（閾値設定手段）では、そのときの発電電流に応じて前記水素ガス質量流量の閾値を設定する。ステップS9では、ステップS7で求めた水素ガス質量流量が、ステップS8で設定した閾値以下であるか否かを判別し、水素ガス質量流量が閾値以下であ

れば、ステップS10（バージ制御手段）へ進んで、バージ弁11を開放してバージを行わせる。

【0044】ところで、定常運転状態では、上記のように、水素ガス質量流量が閾値以下になったときにバージを行わせることで、無用なバージを回避することができるが、特に車両用燃料電池システムでは、燃料電池の負荷変動が大きく、かつ、変動頻度が多い。そして、負荷変動時には、燃料電池の発電電流に対応して設定される閾値の値にもよるが、水素質量流量が不足した状態に入り込みやすくなり、負荷に連動して頻繁にバージ弁11を開放されると、バージ弁開放で排出される水素燃料ガスも無用に増加することになってしまう。

【0045】上記のような過渡的な水素質量流量の不足に対しては、循環系における不純物ガス存在量が少ない\*

$$\text{「不純物ガス存在量」} \propto y \cdot (p/p_0) \cdot (T_0/T) \cdot V \dots (12)$$

【0048】水素質量流量が閾値以下になったときに、上記のようにして求められる不純物ガス存在量に対応する値が閾値以上であるときに、バージを実行させようすれば、負荷変動時に無用にバージが行われることが回避される。尚、水素質量流量のみに基づくバージ制御での図5に示されるバージ領域に対して、圧力、温度を一定と仮定すると、不純物ガス濃度  $y = 1 - \text{水素ガス濃度}$  であるから、上記の水素質量流量及び不純物ガス存在量に基づくバージでは、図7のように不純物ガス濃度の閾値に相当する  $b$  によっても囲まれる斜線領域に示されるより限定した領域でバージが行われることになる。

【0049】図8のフローチャートは、上記の水素質量流量及び不純物ガス存在量に応じたバージ制御を示すものであり、ステップS21～S29の処理は、前記図6のフローチャートのステップS1～S9と同様に行われる。ステップS29（不純物ガス存在量推定手段）で、水素質量流量が閾値以下であると判別されると、ステップS30へ進んで、不純物ガス存在量を演算する。

【0050】ステップS31では、前記不純物ガス存在量が予め記憶された閾値以上であるか否かを判別し、前記不純物ガス存在量が閾値以上であれば、ステップS32へ進んで、バージを行わせる。ところで、図1に示されるシステム構成では、除湿器8の下流側で、水素及び不純物ガスの混合ガスの流量を計測させる構成としたが、循環系の混合ガスが除湿されず、水素、水蒸気、不純物ガスの3成分になっている場合、或いは、図9又は※

$$x = [(1-z) \rho y + z \cdot \rho z - \rho av] / (\rho y - \rho x) \dots (17)$$

$$y = [\rho av - z \cdot \rho z - (1-z) \rho y] / (\rho y - \rho x) \dots (18)$$

となり、水蒸気濃度  $z$  が既知であれば、上式から水素ガス濃度  $x$  及び不純物ガス濃度  $y$  が求められることになる。

【0053】図11は、超音波流量計30が水素、不純物ガス、水蒸気の混合ガスにおける伝播時間を計測する構成において、水蒸気濃度  $z$  を既知として、該水蒸気濃度  $z$  から水素濃度  $x$  を演算するバージ制御を示す。図1

\* 状態では、たとえ上記の水素質量流量不足領域に一時的に在ったとしても、バージ弁を開放しないようにするといい。前記不純物ガス存在量は、還流系の管路等で構成される容積  $V$  を一定と見なせば、式(11)で示される不純物ガス濃度  $y$  と、圧力  $p$  と、温度  $T$  から不純物ガス存在量に対応した量を推定することができる（不純物ガス存在量推定手段）。

【0046】ここで、上記不純物ガス存在量は、流量 ( $L/m^3$ ) ではなく、また、不純物ガス濃度  $y$  そのものと対応しているのではなくて、不純物ガス濃度  $y$  を圧力  $p$  と温度  $T$  で補正した値が存在量に対応することになる。

【0047】

【数1】

$$\text{「不純物ガス存在量」} \propto y \cdot (p/p_0) \cdot (T_0/T) \cdot V \dots (12)$$

※ 図10に示すように、除湿器8の上流側位置でガス流量を検知する場合でも、水蒸気濃度  $z$  を既知とすることで、前記伝播時間  $t_1$ ,  $t_2$ に基づき求められる平均密度  $\rho_{av}$  から水素ガス濃度  $x$  及び不純物ガス濃度  $y$  を求めることができる。

【0051】図9に示す実施形態では、流量計30の上流側近傍に湿度計35を介装させてあり、該湿度計35（湿度検出手段）を備える場合には、該湿度の検出結果と圧力  $p$  とから、水蒸気濃度  $z$  を求めることができる。また、図10に示す実施形態では、湿度計35を備えないが、循環系内が飽和水蒸気状態である場合には、圧力  $p$  と温度  $T$  に基づいて温度  $T$  での飽和水蒸気圧を求めて、水蒸気分圧（濃度）を推定することができる。

【0052】ここで、流量計30は、水素、不純物ガス、水蒸気の混合ガスにおける超音波の伝播時間を計測することになるから、伝播時間  $t_1$ ,  $t_2$ に基づく平均密度  $\rho_{av}$  は、3成分混合ガスの平均密度  $\rho_{av}$  であり、前記式(8), (9)は、水蒸気の密度を  $\rho_z$  とすると、下記のように変更される。

$$x + y + z = 1 \dots (13)$$

$$x \cdot \rho_x + y \cdot \rho_y + z \cdot \rho_z = \rho_{av} \dots (14)$$

ここで、上式を変形して、

$$x + y = 1 - z \dots (15)$$

$$x \cdot \rho_x + y \cdot \rho_y = \rho_{av} - z \cdot \rho_z \dots (16)$$

として、これを未知数  $x$ ,  $y$ について解くと、

$$x = [(1-z) \rho y + z \cdot \rho z - \rho_{av}] / (\rho y - \rho x) \dots (17)$$

$$y = [\rho_{av} - z \cdot \rho z - (1-z) \rho y] / (\rho y - \rho x) \dots (18)$$

1に示すフローチャートは、ステップS25で平均密度  $\rho_{av}$  を演算すると、次のステップS26A（濃度推定手段）では、湿度計35に基づき、或いは、循環系内が飽和水蒸気状態である場合に圧力・温度から、水蒸気濃度  $z$  を演算する。

【0054】そして、ステップS26Bでは、前記水蒸気濃度  $z$  を用いて水素ガス濃度  $x$  を演算する。上記ステ

ップS26A、ステップS26B以外の各ステップでは、図8と同様な処理が行われる。尚、循環系内における湿度、圧力、温度が略一定である場合には、湿度計35、圧力計36、温度計37を設けなくても、水蒸気濃度 $z$ を予め一定値と見なして、水素ガス濃度 $x$ 及び不純物ガス濃度 $y$ を求めることが可能である。

【0055】また、流量計30を図12に示すように、水素ガス循環系を構成する供給ライン5に介装させる構成としても良い。この場合、計測される水素質量流量は、エジェクタ4によって還流される分と、燃料電池スタック20における消費分に対応する供給ライン3からの供給分との合計になるから、発電電流に応じて設定する水素質量流量の閾値を、供給ライン3からの供給分だけ嵩上げすれば、前記実施形態と同様にしてバージ制御を行わせることができる。

#### 【図面の簡単な説明】

【図1】除湿器の下流側に流量計を備える実施形態のシステム構成図。

【図2】超音波流量計の伝播時間と諸物理量の関係を説明するための模式図。

【図3】音速演算値と混合ガスの平均密度との関係を示す線図。

【図4】水素質量流量と濃度と混合ガス流量の関係を表す線図。

【図5】定常運転状態での水素質量流量不足領域を表す線図。

【図6】水素質量流量に基づくバージ制御を示すフローチャート。

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\* 【図7】水素質量流量と不純物ガス存在量に基づくバージ領域を表す線図。

【図8】水素質量流量と不純物ガス存在量に基づくバージ制御を示すフローチャート。

【図9】湿度計を備える実施形態のシステム構成図。

【図10】湿度計を備えない実施形態のシステム構成図。

【図11】水蒸気濃度から水素ガス濃度を求めるバージ制御を示すフローチャート。

10 【図12】流量計を燃料ガス供給系に配置した実施形態を示すシステム構成図。

#### 【符号の説明】

1…水素ガス供給源

2…供給調圧バルブ

3…水素ガス供給ライン

4…エジェクタ

5…水素ガス供給ライン

6…加湿器

7, 9, 10…水素ガス還流路

8…除湿器

11…バージ弁

12…排出ライン

30…流量計

35…湿度計

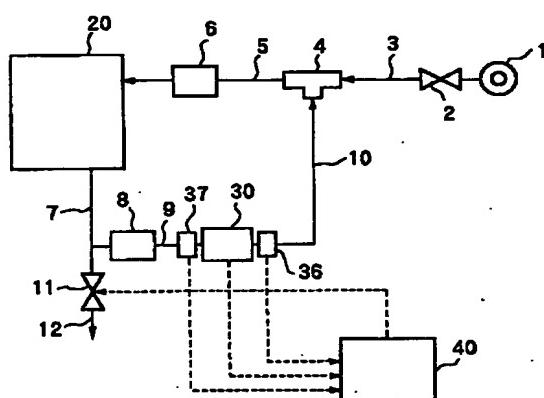
36…圧力計

37…温度計

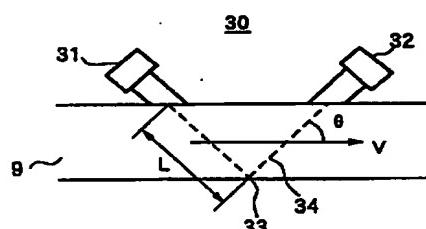
40…信号演算処理部

\*

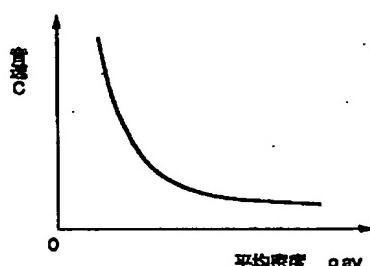
【図1】



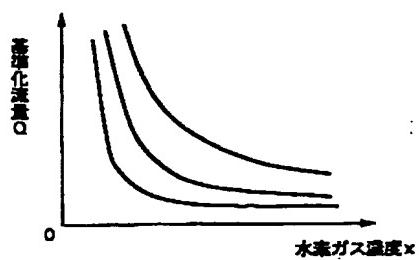
【図2】



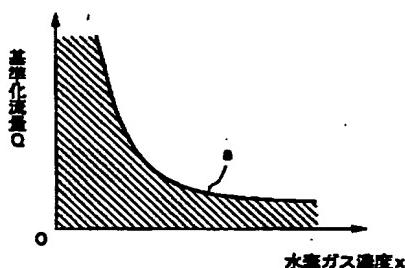
【図3】



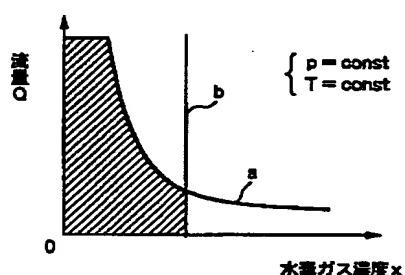
【図4】



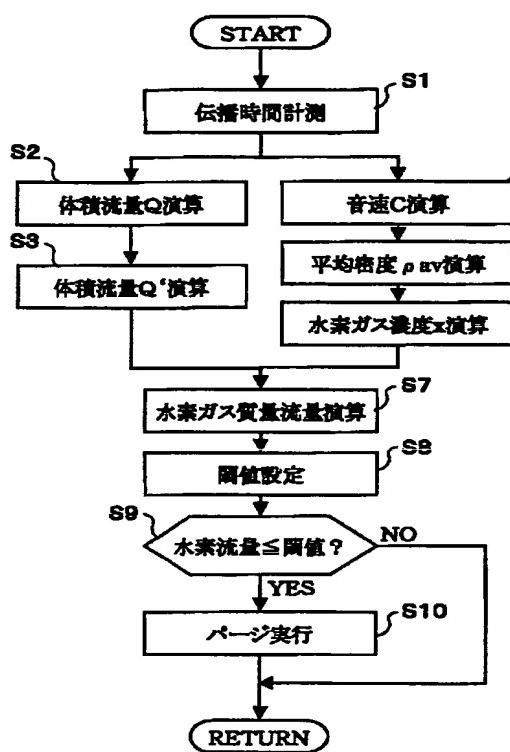
【図5】



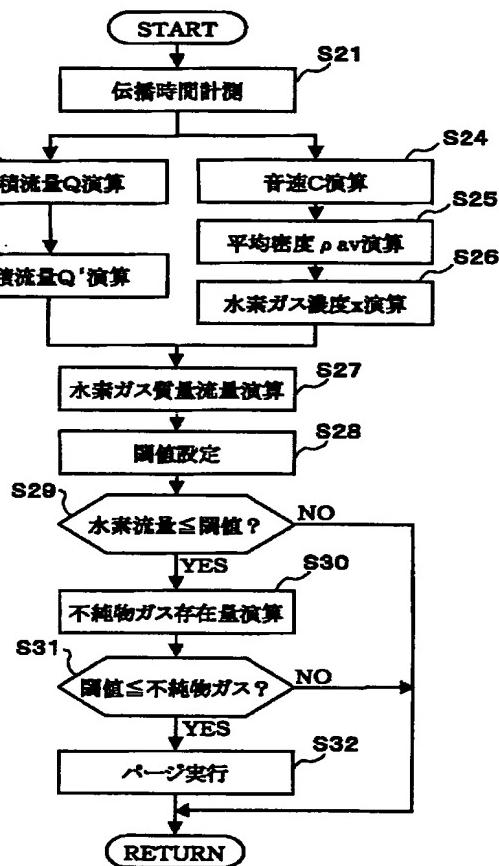
【図7】



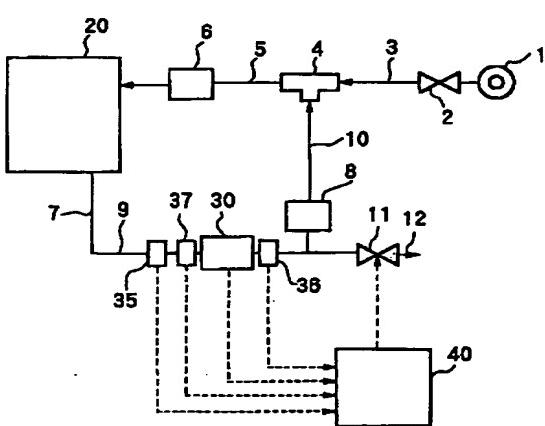
【図6】



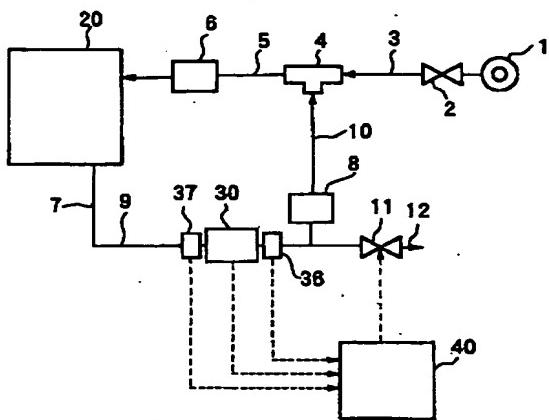
【図8】



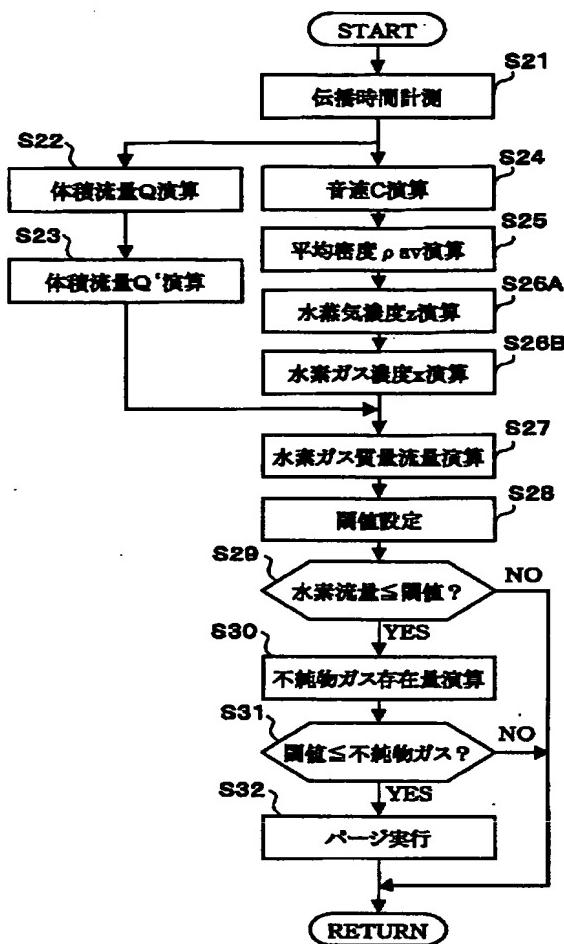
【図9】



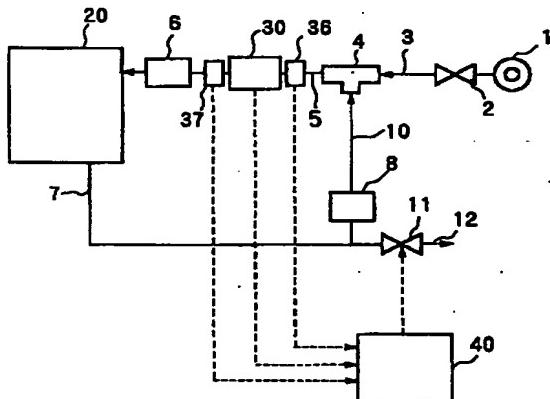
【図10】



【図11】



【図12】



フロントページの続き

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KK31 KK44 KK56 MM08